

Determining surface magnetization and local magnetic moments with atomic scale resolution.

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We propose a method to determine the direction of surface magnetization and local magnetic moments on the atomic scale. The method comprises high resolution scanning tunneling microscope experiments in conjunction with first principles simulations of the tunneling current. The potential of the method is demonstrated on a model system, antiferromagnetic Mn overlayers on W(110). We expect that it will ultimately allow to study the detailed changes of magnetic surface structures in the vicinity of dopants or impurities.

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Until very recently magnetic properties of thin metal films or surfaces could be determined only by x-ray magnetic dichroism experiments [1–3]. In these experiments x-rays of circular polarization, originating from synchrotron radiation, are used to probe the magnetic properties. With the help of second order perturbation theory the intensity of the adsorbed radiation can be related to magnetic moments μ and the direction of spin polarization \vec{M} due to crystal anisotropies [4]. However, the method suffers from two serious weaknesses, which limit its applicability: (i) The local resolution obtained with synchrotron radiation is far too low for any atomic scale analysis and in the range of 50nm [5]. (ii) The intensity of the x-ray beams is high enough to lead to substantial energy dissipation. Since magnetic properties are very sensitive to temperature changes, the measurement of groundstate properties is problematic.

A solution to these problems could come from a different method, one that does not affect the electronic states of a magnetic layer in any substantial way. The spin polarized (SP) scanning tunneling microscope (STM) does provide just such a method. In measurements with an iron coated tungsten tip on single antiferromagnetic Mn layers it has been demonstrated that the local magnetic moment can be resolved on the atomic scale by STM scans [6]. The theoretical simulation of these scans revealed that such a scan is very sensitive to the chemical nature of the STM tip apex, which allows one to identify the STM tip from the corrugation height of the surface [7]. However, these results were obtained under the condition of ferromagnetic ordering of sample and tip states, the direction of surface magnetization was therefore imposed from the outset.

In this Letter we propose an extension of the method to account for truly general orientations of the magnetic axis. We want to show, how the orientation of the magnetization vector \vec{M} of sample and tip changes the tunneling current and the corrugation height measured on a surface. This in turn allows one to determine the magnetic axis and the local moments from STM scans and first principles simulations. Since antiferromagnetic surface

ordering on the atomic scale has already been observed [6], it is justified to assume that magnetic surface properties are not decisively modified by an STM tip. And as the experiments are suitable to reveal electronic properties on an atomic scale, they also remove the resolution problem one is confronted with in dichroism measurements.

Let us consider the situation in a tunnelling junction between a crystal surface and an STM tip in real space. Magnetic anisotropy in a crystal breaks the rotational symmetry of electron spins. The spin states in this case are projected onto the crystal's magnetic axis. We assume in the following that this symmetry breaking occurs in the two separate systems which form our tunnelling junction. Depending on the orientation of the magnetic axes two limiting cases have to be distinguished. The magnetic axis of sample and tip are either parallel or antiparallel. In the first case we have to sum up all electrons tunnelling from spin-up states of the sample (n_S^\uparrow) to spin-up states of the tip (n_T^\uparrow). This ferromagnetic ordering is described by the following transitions:

$$\mathbf{M}_S \uparrow \quad \left\{ \begin{array}{l} n_S^\uparrow \longrightarrow n_T^\uparrow \\ n_S^\downarrow \longrightarrow n_T^\downarrow \end{array} \right\} \quad \uparrow \mathbf{M}_T \quad (1)$$

Here \mathbf{M}_S and \mathbf{M}_T describe the magnetic axes of sample and tip, respectively. We denote the tunnelling current due to ferromagnetic ordering by I_F . If the two vectors are antiparallel the electrons tunnel from states (n_S^\uparrow) into states (n_T^\downarrow) and vice versa. The antiferromagnetic ordering is therefore described by:

$$\mathbf{M}_S \uparrow \quad \left\{ \begin{array}{l} n_S^\uparrow \longrightarrow n_T^\downarrow \\ n_S^\downarrow \longrightarrow n_T^\uparrow \end{array} \right\} \quad \downarrow \mathbf{M}_T \quad (2)$$

This setup yields the antiferromagnetic tunnelling current I_A . Since the energy of the tunnelling electrons is very low, and since the overlap of the sample and tip wavefunctions is computed far outside the core region of surface atoms, spin-orbit coupling can generally be neglected in the theoretical treatment. Within density functional theory (DFT) [8,9] the tunnelling current is

commonly described in terms of ϕ_M , the angle between the two magnetic axes, and $P_{S(T)}$, the polarization of the sample (tip) surface:

$$I(\phi_M) = I_0(1 + P_S P_T \cos \phi_M) \quad (3)$$

$$\cos \phi_M = \frac{\mathbf{M}_S \cdot \mathbf{M}_T}{|\mathbf{M}_S||\mathbf{M}_T|} \quad (4)$$

For constant tunnelling matrix elements and within a perturbation approach the current I_0 and polarizations $P_{S(T)}$ are given by:

$$I_0 \propto \frac{1}{2} (n_S^\uparrow + n_S^\downarrow) (n_T^\uparrow + n_T^\downarrow) \quad (5)$$

$$P_{S(T)} = \frac{n_{S(T)}^\uparrow - n_{S(T)}^\downarrow}{n_{S(T)}^\uparrow + n_{S(T)}^\downarrow} \quad (6)$$

I_0 and $P_S P_T$ can be written in terms of the ferromagnetic and antiferromagnetic currents:

$$I_0 = \frac{1}{2} (I_F + I_A) \quad P_S P_T = \frac{I_F - I_A}{I_F + I_A} \quad (7)$$

If the tunnelling matrix element is not constant, the current has to be calculated numerically from the Bardeen integral over the separation surface [10,11]. In this case the current contributions have to account for the spin orientation of a given eigenstate. In DFT the energetic minimum for magnetic crystals is reached by optimizing the distribution of spin-up density n^\uparrow and spin-down density n^\downarrow . The currents for ferromagnetic and antiferromagnetic coupling are computed by calculating the transition matrix elements for the spin polarized Kohn-Sham states of sample and tip:

$$I_F = I(n_S^\uparrow \rightarrow n_T^\uparrow) + I(n_S^\downarrow \rightarrow n_T^\downarrow) \quad (8)$$

$$I_A = I(n_S^\uparrow \rightarrow n_T^\downarrow) + I(n_S^\downarrow \rightarrow n_T^\uparrow) \quad (9)$$

Calculating the tunnel current for different angles ϕ_M requires then only to compute the linear combination of ferromagnetic and antiferromagnetic currents multiplied by the appropriate coefficients. From these three dimensional current maps the constant current contours and the surface corrugations can be extracted in a straightforward manner.

We have used a full potential method to compute the electronic groundstate properties of model tips. The tip in the experiments was a tungsten wire coated with several layers of iron [6]. We mimic this tip by an ideal Fe(100) surface with a single Fe atom in the apex (see Fig. 1). Previous simulations of STM experiments revealed that the best agreement between experiments and simulations is often obtained with a tip model, covered by impurities of the sample surface [11]. Therefore we

modified the clean Fe-tip by two additional setups. In one case the apex atom was changed to Mn, in the second also the surface layer consisted of Mn atoms (see Fig. 1). On the technical side we note that the free standing film consisted of five Fe layers and two additional layers for the apex. 10 special k-points were used in the spin polarized DFT calculations. Given the $c(2 \times 2)$ unit cell this amounts to 40 k-points for the elementary cell. The convergence in the final iterations was better than 0.01 e/au³. Since Mn and Fe are both 3d metals and relaxations are therefore rather small, the STM simulations were based on the wavefunctions of bulk truncated crystals. The details of the electronic structure calculations are published in a separate paper [12].

In the Bardeen approach to tunnelling [10] the tunnelling current is computed by integrating the overlap of sample and tip states over the separation surface. In our program, which is described in detail elsewhere (bSCAN [11]), we integrate over a finite separation area and sum up all contributions from the Kohn-Sham states of sample and tip numerically. It has been shown previously that the STM current in the experiments on W(110)Mn is about one to two orders of magnitude higher than the current obtainable within a perturbation approach and at a reasonable distance [7]. Even though we could not pin down this discrepancy, it seems to be most likely due to current leakages of the STM circuit. In our implementation of perturbation theory it is implicitly assumed that all tunnelling current passes through a small area of the separation surface, thus it cannot account for off center contributions in the circuit. Therefore we computed the constant current contours not for the actual values in the measurements (which were in the range of 30nA [6]), but chose contours centered at about 4.5 to 4.6 Å above the W(110)Mn film. This distance is at the lower limit of mechanical stability on metal surfaces [13]. The bias voltage in the measurements was - 3 mV, we chose the same voltage for our simulations. In the figures we display the constant current contours depending on two separate parameters, important for every single STM experiment: (i) The chemical nature of the tip surface. We report on simulations with three different chemical compositions of the tip. (ii) The angle of magnetization. We show simulations for selected angles ϕ_M between the magnetic axis of the crystal surface and the STM tip.

The electronic structure of the W(110)Mn surface in antiferromagnetic ordering depends substantially on the spin orientation of electron charge. For spin-up states the density contours have a maximum at the position of Mn atoms with a negative magnetic moment and their minimum at the position of atoms with the opposite polarization (see Fig. 2). For spin-down states the situation is reversed: atoms with positive magnetic moments are now seen as protrusions. This indicates that the surface has the highest corrugation for charge transport with a high degree of polarization. In addition, it shows that the surface is comparatively flat if measured by a paramagnetic tip (see Fig. 2, right frame).

Figs. 3 to 5 display the results of our STM simulations with clean and Mn contaminated Fe model tips. We show five angular settings, from $\phi_M = 0$ (ferromagnetic charge transitions) to $\phi_M = 180^\circ$ (antiferromagnetic charge transitions). In general the obtained magnetic contrast depends on the angle ϕ_M . It is a maximum for the limiting values of 0° and 180° , and it vanishes for $\phi_M = 90^\circ$. This angle denotes the case where tunnelling transitions from the sample into the tip are essentially unpolarized. We note two distinct features in our simulations:

- The obtained maximum of magnetic contrast depends on the chemical nature of the tip apex.

It is 68pm for a clean tip; 89pm for a tip contaminated with an Mn atom; and it nearly vanishes for a tip contaminated by a surface layer of Mn (4pm, Fig. 5). We may therefore conclude that high resolution measurements with a suitable degree of precision, measurements which are already possible at present, allow to differentiate between different tips also in case of a magnetic tunnel junction. For non-magnetic tunnel junctions, we have already shown the influence of the tip in previous publications [11]. Comparing the simulations with the experiments [6], the most likely tip in the actual experiments seems to be tip model (c) (see Fig. 1): in this case the corrugation is within the range of experiments over a wide range of angles (the range is about 2-4pm and it varies, seemingly due to the lateral angle of surface polarization [14]). This indicates that the STM tip during the measurement is contaminated by at least a monolayer of surface atoms; we would conclude from this feature that the tip is generally very close to the surface and that the experiments are indeed done at the lower limit of stability [13].

- In general the measured magnetic contrast depends on the angle ϕ_M between tip and sample magnetization.

Therefore this angle can be uniquely determined by comparing experimental results with simulations. The experimentally obtained magnetic contrast was 2 to 4pm [14]. In our simulations this value indicates either, that the tip was covered by at least one monolayer of Mn, or that the angle between the two magnetization vectors was $87 - 89^\circ$ (Fe tip apex or Mn tip apex). Given that this seems highly improbable for two coplanar vectors, the most likely explanation of the experiments is that they were performed with a tip close to our model (c) (see Fig. 1). However, this tip is the least suitable to resolve the angle ϕ_M . With a clean tip, or a tip only contaminated by single atoms of the sample, this angle could be resolved accurately to about $1-2^\circ$.

The last step in obtaining all magnetic properties on the atomic scale is the variation of the angle ϕ_M . This can essentially be done in two ways: (i) Either the sample or the STM tip can be rotated. (ii) The orientation of the

magnetic field of the STM tip is stabilized by an external magnetic field, and this external magnetic field is rotated. In both cases images have to be recorded for a number of settings. From these images the direction in space of the sample magnetization can be uniquely determined.

The method has a number of important implications. First, it can lead to a detailed understanding, how magnetism depends on the layer structure of a magnetic multilayer. As the controlled growth of single layers is experimentally feasible, magnetic properties can be directly studied, which change with the number of layers, e.g. the orientation of the magnetization axis [2]. Second, it is possible to study the influence of impurities on the local magnetic field. Given that magnetic properties have such a wide range of applications in current technology, this possibility to study the site-dependency of magnetism on the atomic scale could lead to a much better understanding of the chemical determinants of magnetic systems. We think that this potential alone makes the method, and the SP STM, the most promising tool in nanomagnetic research.

In summary we have shown that the combination of high resolution spin-polarized STM scans and first principles simulations of the tunnel current makes it possible to determine all magnetic properties of a surface on the atomic scale: the local magnetic moment as well as the polarization and the direction of surface magnetization. We expect that this method will ultimately allow to study changes of the magnetic surface properties in the vicinity of dopants and impurities.

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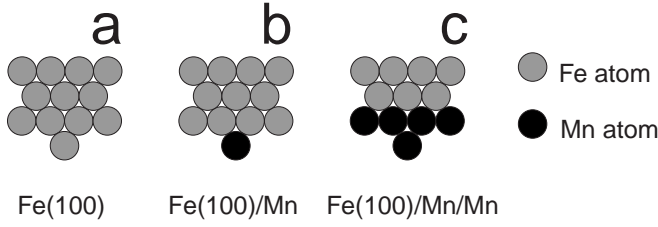


FIG. 1. STM tip models in our simulation. The tip is mimicked by a $c(2 \times 2)$ Fe(100) free standing film with a single apex atom (a). Contamination of the tip by atoms of the crystal surface is accounted for by a single Mn apex atom (b), or an Mn monolayer and a Mn apex atom (c).

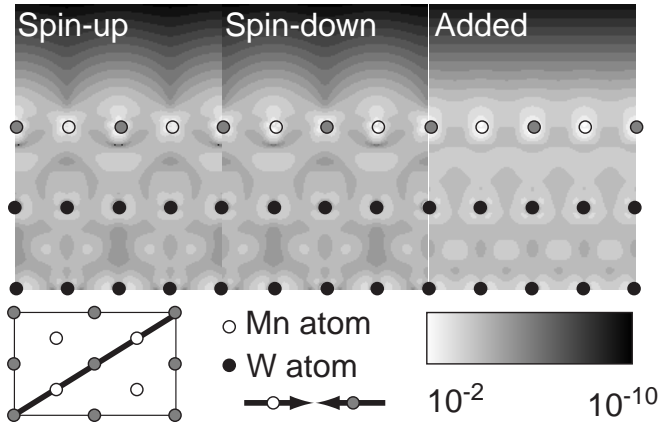


FIG. 2. Electronic structure of the W(110)Mn film. The plots show the local density of states in a vertical plane cut through the crystal (sketch at the bottom, left). The magnetic moment in the surface atoms is either positive (empty circles), or negative (grey circles). The LDOS above different atoms reveals these atoms either as protrusions, or as depressions, depending on the electron spin. The corrugation of the surface layers vanishes for paramagnetic STM tips.

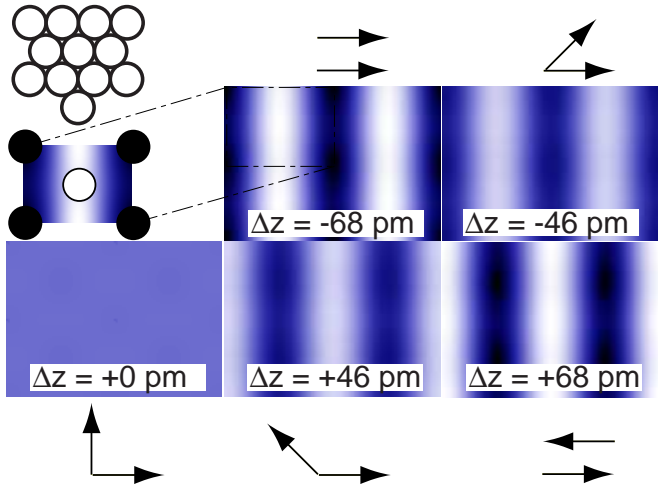


FIG. 3. Simulated STM scans for a Fe(100) tip with Fe apex. The simulations show four unit cells, the orientation of the magnetic axes for the individual scans is depicted by the black arrows. The corrugation height Δz is the difference in apparent height between atoms of positive (full circles) and negative (empty circle) magnetic moments. For perpendicular moments the surface corrugation vanishes.

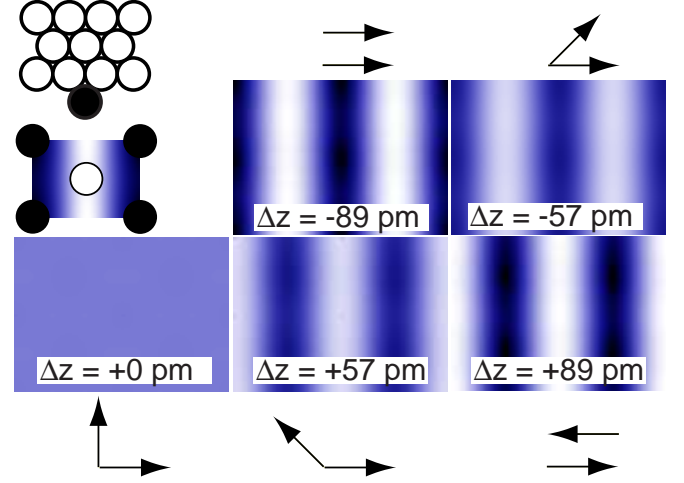


FIG. 4. Simulated STM scans for a Fe(100) tip with Mn apex. The corrugation in this case is higher, the apparent height is greater for surface atoms with a positive magnetic moment.

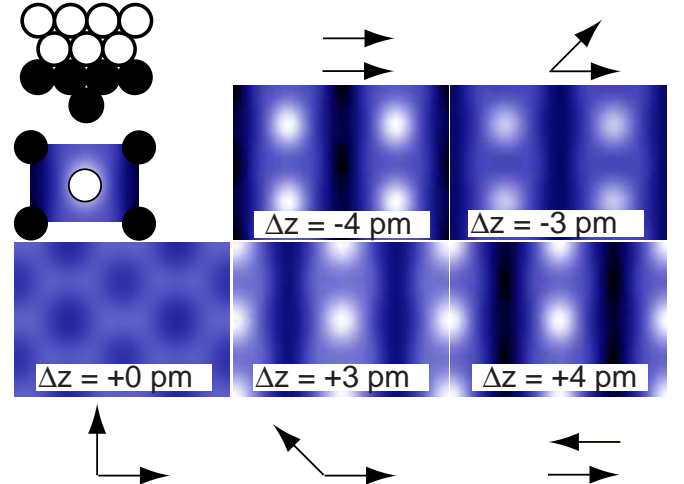


FIG. 5. Simulated STM scans for a Fe(100)Mn tip with Mn apex. The magnetic contrast in this case nearly vanishes. The corrugation height remains unchanged over a wide range of angles, contrary to the simulations with clean or moderately contaminated STM tips.